Thermoplastic Electroactive Gels for 3D-printable Artificial Muscles

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Abstract

3D-printable artificial muscles have only recently garnered interest, and few 3D-printed artificial muscles have been demonstrated. In this article, we introduce the concept of thermoplastic electroactive gels, new smart materials that can be fabricated simply and rapidly by heating, overcoming the limitations of previous dangerous and time-consuming solvent-based manufacturing methods, and enabling hot-pressing, melt-recycling, extrusion and 3D-printing. We present and characterise a new example material, PVC-DIDA (polyvinyl chloride and diisodecyl adipate) gel and demonstrate multi-layer PVC-DIDA gel artificial muscles. Finally, the extrudability of PVC-DIDA gel is confirmed, and an artificial muscle made from extruded electroactive gel is presented. The electroactivity and extrudability of these thermoplastic gels highlights them as excellent candidate materials for 3D-printing artificial muscle structures.

Keywords: artificial muscles, PVC gel, actuators, soft robotics, 3D printing

Introduction

High-performance artificial muscles are needed for the next generation of medical, industrial and robotics applications [1]. They could replace failing biological muscle, outperform conventional actuators in many machines and power novel robots [2]. In particular, electroactive polymers have the advantage that their power source (electricity) is readily available, in contrast to other artificial muscles such as hydraulic or pneumatic devices, which require large, heavy compressors or pressure vessels. Notable electroactive polymers for artificial muscles include dielectric elastomer actuators [3], ionic-polymer metal composites [4] and electroactive gels, such as hydrogel-based [5], polyurethane gel [6] and PVC (polyvinyl chloride) gel actuators [7].

Gels, which are materials composed of liquid particles dispersed in a solid network, are attractive candidate materials for artificial muscles due to their inherent softness and elasticity [8], vast range of available materials and the potential for self-healing behaviour [9]. 3D-printable artificial muscles have only recently garnered substantial interest [10], and there are only a few examples, such as 3D-printed fluidic actuators [11,12], fluidically actuated soft robots [13] and dielectric elastomer structures [14–17].

PVC gel actuators are a recent type of electroactive smart material [18]. When PVC gel is placed between two electrodes, it exhibits unusual “anodophilic” (anode-loving) behaviour, creeping towards the anode and increasing its anode-contacting surface area (Figure 1(a), Video S1)

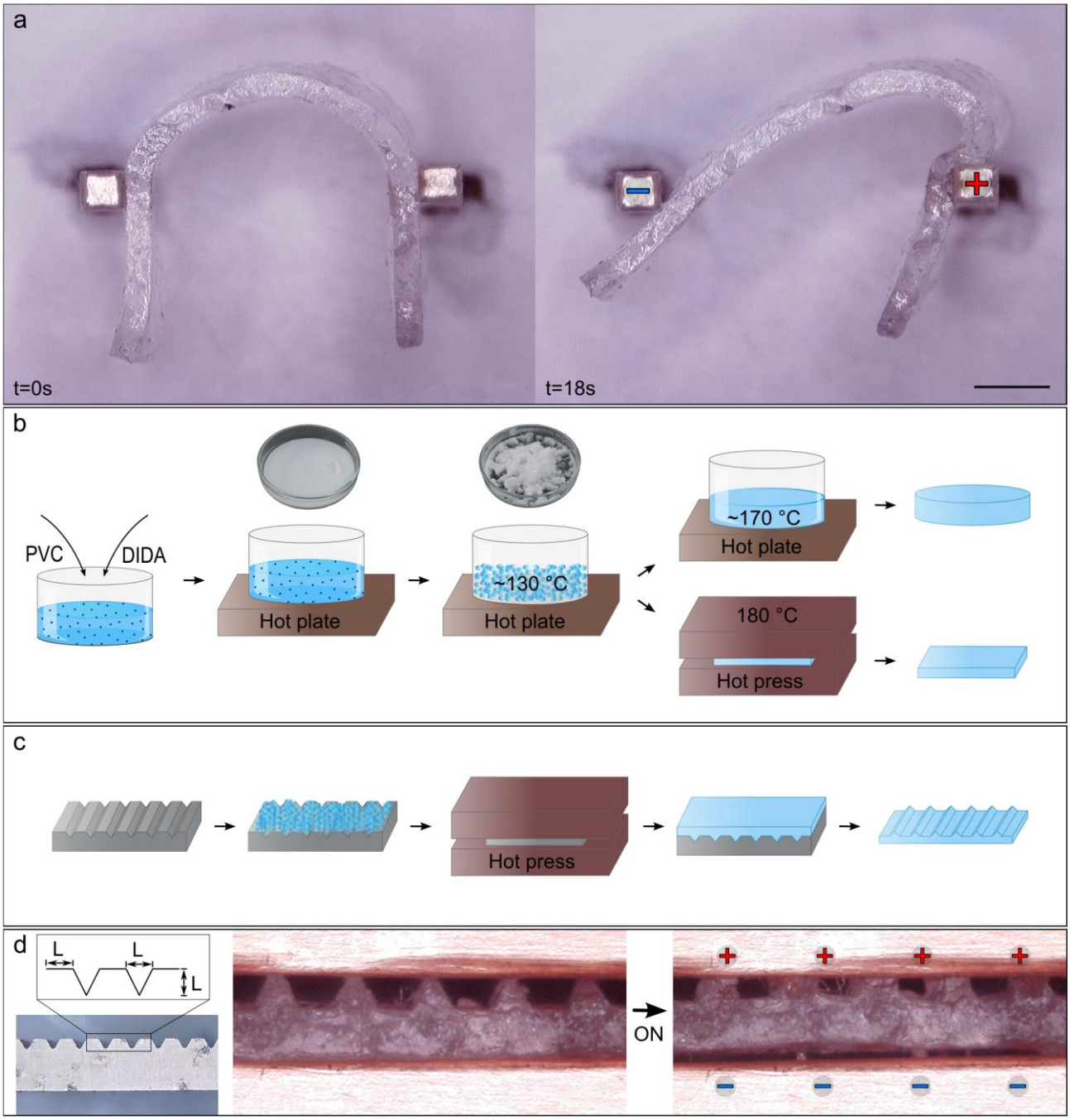

Figure 1. PVC gel behaviour, and thermoplastic PVC gel manufacture and testing. (a) Anodophilic PVC gel behaviour. When the electrodes are charged, the PVC gel crawls towards the anode, increasing its anode-contacting surface area. Scale bar indicates 1 mm. (b) Manufacture of thermoplastic PVC-DIDA gels. (c) Manufacture of microstructured PVC-DIDA gels. (d) Negative mould used to manufacture microstructured PVC-DIDA gels, and anodophilic behaviour of the resultant microstructured gel. L = 0.5 mm.

[7,18,19]. For PVC gel actuators made using dibutyl adipate (DBA), this behaviour has been attributed to "migration of [the] plasticizer-rich phase" of the gel, evidenced by the observation of a softer (higher plasticiser content) local layer adjacent to the anode [19–21]. This anodophilic behaviour can be exploited as an actuation mechanism to achieve bending [18] or contraction [22].

More unusual PVC gel arrangements include an actuator based on wave-shaped gels [23], textile and yarn actuators [24] and an entirely soft actuator using surface-microstructured gels [25]. However, in all these cases, the

constituent PVC gels were fabricated using a dangerous and time-consuming solvent-based method.

In this article, we present a new concept as a step towards 3D-printable gel artificial muscles: thermoplastic electroactive gels. We present the first example of a meltable electroactive gel, a new material to add to the anodophilic smart material family: PVC-DIDA (polyvinyl chloride and diisodecyl adipate) gel. Its meltability implies several significant advantages compared with previous PVC gel actuators. Fabrication can be performed in minutes, simply by heating, which compares favourably with previous multi-day fabrication methods using dangerous solvents. Since these new electroactive gel materials can be melted, they can be formed and reformed into sophisticated smart structures that are not possible using traditional solvent-based fabrication methods. Finally, the potentiality of extrusion makes these smart gels ideal candidates for additive layer manufacturing, laying the foundation for 3D-printable electroactive gel artificial muscles.

Methods

In the traditional PVC gel actuator manufacturing method, PVC and a plasticiser are dissolved in a solvent—typically THF (tetrahydrofuran, which is flammable, toxic and carcinogenic) [7,18,22]. The resulting solution is left at room temperature for between several days [26,27] and 1 week [18] for the THF to evaporate, leaving behind a soft, compliant gel. In addition to being dangerous and time-consuming, this fabrication process limits the range of structures that may be produced.

In contrast, plasticisers have been used to make PVC soft and flexible since the 1920s [28], and the normal manufacturing method for plasticised PVC requires no solvent: a dispersion of plasticiser and PVC particles is simply cured at 190–200 °C [29]. The pervasiveness of the solvent-based method for manufacturing PVC gel actuators may stem from the fact that the anodophilic PVC gel has almost always been manufactured using DBA plasticiser [7,20–22,24,26,30–32], whose flash point is 113 °C [33], whereas rigid PVC melts at about 170–180°C [34]. This has prevented heat-based manufacture and necessitated the use of solvents. In contrast, in this article, we propose the use of high-flash-point, high-boiling-point plasticisers to achieve electroactive behaviour.

As a proof of concept, we demonstrate a new anodophilic material composed of PVC and DIDA (diisodecyl adipate), a plasticiser with a flash point of 213 °C and boiling point of 349 °C [35]. To create the PVC gel, the plasticiser and PVC are first mixed well to form a plastisol. Within the plastisol, the PVC particles absorb some of the plasticiser at room temperature [29]. The plastisol is gently heated to around 130 °C, which accelerates the absorption of plasticiser by the PVC, and the plastisol transitions to a wet crumbly paste (Figure 1(b)). This crumbly paste then acts as a direct precursor to PVC gel; it can be heated to around 170 °C, at which point the plasticised PVC particles fuse together to form a homogenous PVC gel. Alternatively, the plastisol can be hot pressed to form thin PVC gel structures. The resultant PVC gel exhibits the same anodophilic behavior as previously studied in PVC-DBA gels, and we assume it is driven by a similar mechanism. The exact mechanism driving electroactivity in PVC-DIDA gels will be the topic of future study.

In typical contracting PVC gel actuators, a thin sample of PVC gel creeps into the porous structure of a mesh-like anode, delivering an overall reduction in thickness. In contrast, previously demonstrated surface-microstructured PVC gels incorporate the microstructure required for bulk contraction directly on their surface [25]. Using the new concept of thermoplastic electroactive gels, hot pressing allows easy fabrication of high-quality microstructured PVC gels: any microstructure that lacks overhangs can be produced by hot pressing plastisol paste onto a negative mould (Figure 1(c)). In this research, we used a standard V-shaped mould with a feature length (L) of 0.5 mm (Figure 1(d)). Hot pressing using this mould structure produces a gel with triangular microstructures, which have been shown to maximise anodophilic-behaviour-driven actuation compared with square, trapezium, and U-shaped microstructures in PVC-DBA electroactive gels [25]. These triangular microstructures allow a large amount of material to migrate to the anode, resulting in considerable overall PVC gel movement (Figure 1(d), Video S2).

Results and discussion

The electromechanical properties of the microstructured PVC-DIDA gel are shown in Figure 2. Two standard test rigs were used to record isometric and isotonic behaviour (Figure 2(a) and 2(b)). In all experiments described in this article, the microstructured PVC-DIDA gel sample total thickness (including microstructure height) was 1 mm. The initial passive stiffness of microstructured PVC-DIDA gels at various plasticiser mass ratios is shown in Figure 2(c); higher concentrations of DIDA resulted in softer gels.

During testing, a known potential difference (for example, Figure 2(d) shows 800 V) was applied between the electrodes for 10 to 60 seconds (depending on mass ratio) to capture full actuation. Actuation and relaxation were asymptotic, with all samples reaching 80% of full stroke within 5 to 10 seconds, and 80% of full relaxation within 5 to 20 seconds. Noteworthy actuation only occurred when a positive voltage was applied; when a negative voltage was applied, actuation was negligible (Fig. 2(d)), since the gel microstructure was adjacent to the cathode rather than the anode and so anodophilic creep did not contribute to contraction. This asymmetry confirms that contraction was caused by anodophilic creep rather than simple gel compression by Maxwell's stress, which is responsible for the contraction of dielectric elastomer

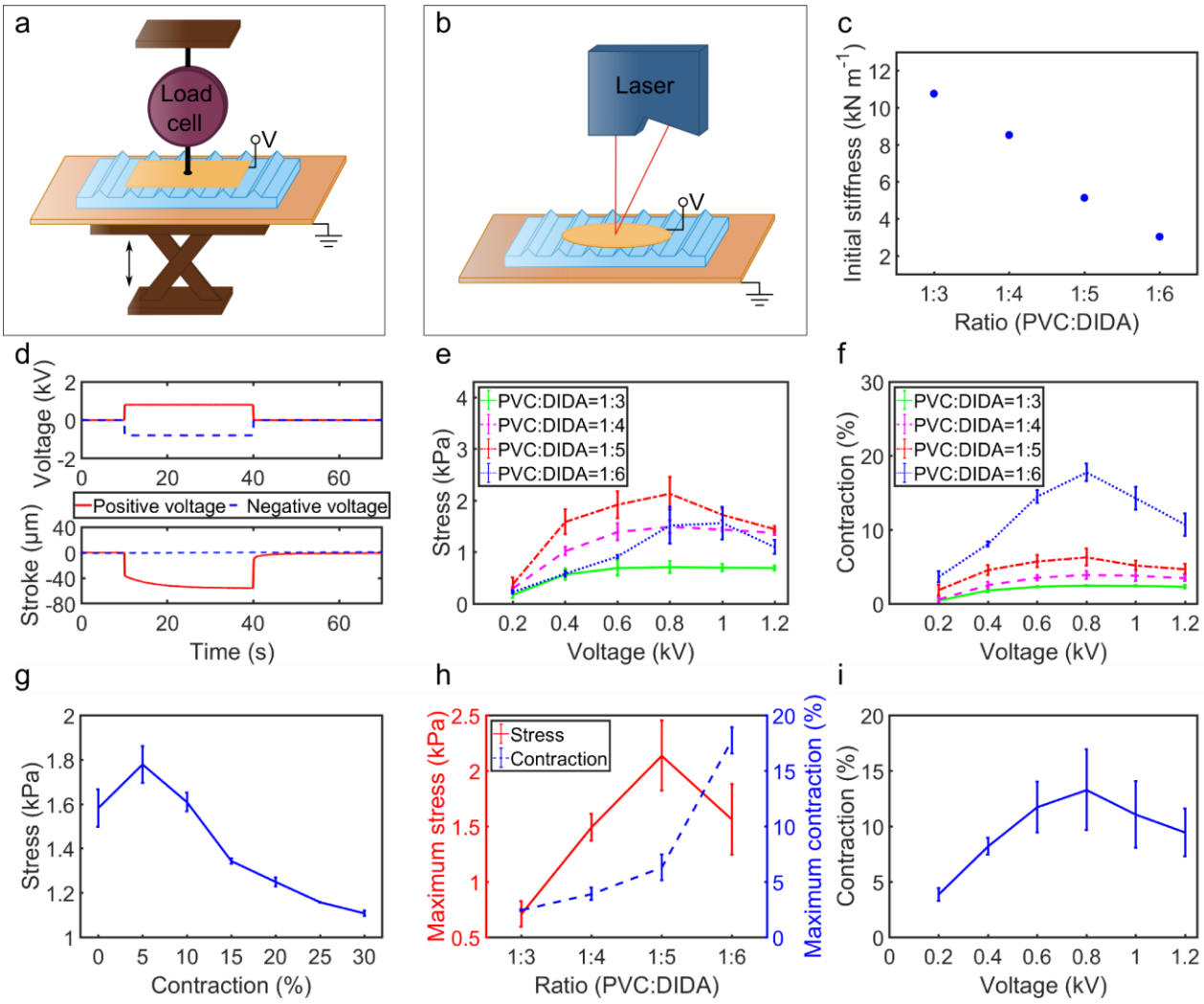

Figure 2. Testing and properties of microstructured PVC-DIDA gels. (a) Isometric testing setup. (b) Isotonic testing setup. (c) Initial stiffness of microstructured gels as plasticiser mass ratio is varied. (d) Example voltage and stroke during testing. (e) Actuation stress as voltage is varied for various plasticiser ratio gel samples. (f) Contraction as voltage is varied for various plasticiser ratio gel samples. (g) Stress variation with contraction for a 1:6 ratio gel under 800 V. (h) Maximum stress and maximum contraction as plasticiser ratio is varied. In (e)-(h), points are averages of three trials and error bars show ± one standard deviation between trials. (i) Sample variance between samples of 1:6 ratio gels. Points are averages of three samples and error bars show ± one standard deviation between samples.

actuators [3], and occurs irrespective of polarity. Additionally, for the gels presented in this article the voltage required to achieve noteworthy actuation (less than 1 MVm^{-1}) is extremely low compared with that of dielectric elastomer actuators (more than 50 MVm^{-1}) [3].

Figure 2(e) and 2(f) show results from isometric and isotonic testing of microstructured PVC-DIDA gels. As can be seen from Figure 1(d), PVC gels migrate towards and adhere to the anode but do not exert a strong attractive force on the cathode [20]; they may even migrate away from it. In the past, this has made the measurement of contractile force challenging, leading researchers to measure the force on a cathode entirely encapsulated in PVC gel [22], measure only the adhesive force of the gel on the anode [20], or report "recovery" properties; the force or stress exerted by a PVC gel actuator as it relaxes after voltage is removed [31,32]. The meltability of thermoplastic electroactive gels allows for the measurement of the true contractile force of gel samples: for each sample of PVC-DIDA gel, a small quantity of gel was melted onto the surface of an aluminium cathode and used as an adhesive to firmly attach the sample to the cathode. Maximum actuation stress was 2.137 kPa and maximum contraction 17.75%.

Typically, both stress and contraction were maximised when around 800 V (field strength 0.8 MVm^{-1}) was applied. The reasons for reduction in stress and contraction above 800

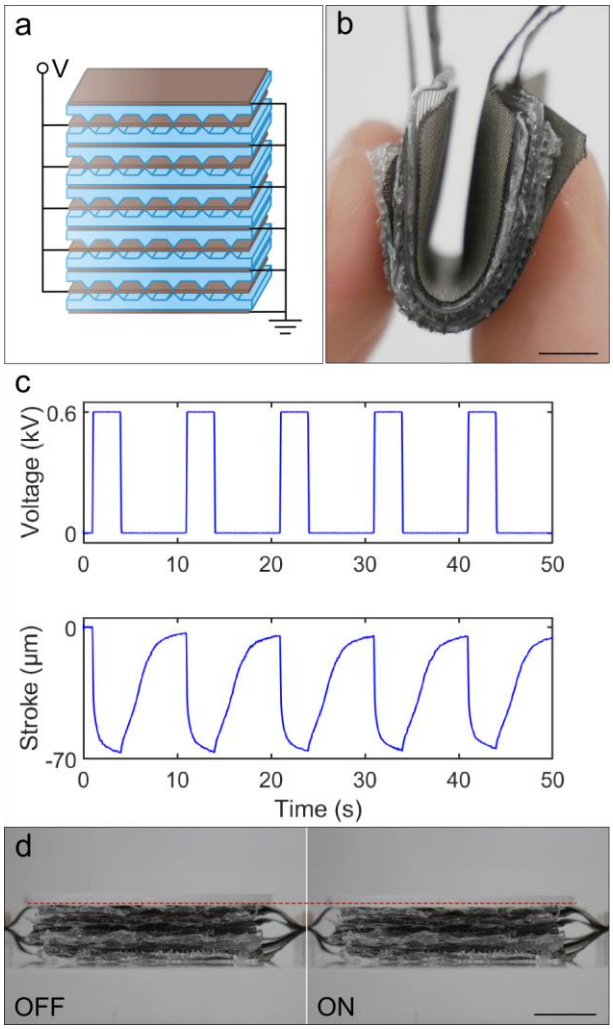

Figure 3. PVC-DIDA gel artificial muscles (a) Ten-gel-layer PVC-DIDA gel artificial muscle structure. (b) Photo showing flexibility of four-gel-layer artificial muscle. (c) Cyclic test of three-gel-layer PVC-DIDA artificial muscle (d) 5.5 % contraction of ten-gel-layer PVC-DIDA artificial muscle. Scale bars indicate 1 cm.

V warrant further investigation. We observed that up to 800 V, steady-state current increased proportionally with voltage, however above this point current increased more quickly, indicating a reduction in steady-state resistance. Based upon this observation, one explanation is that above 800 V, the higher conductance of the gel reduced the actual electrical field that could be sustained and thus actuation was weaker. For example, the gel might be exhibiting partial discharge [36]. Local leak-induced heating could also contribute to this partial breakdown [37,38].

Figure 2(g) shows exerted stress when 800 V was applied at various levels of contraction. The highest stress was achieved by slightly compressing the gel (by 50 μm), which resulted in a greater stress than when the gel was just touching (but not compressed by) the anode. We propose that slightly compressing the gel guarantees a good contact between each microstructure tip and the anode, which ensures anodophilic creep occurs at every microstructure, maximising contractile stress. Increasing compression beyond this point appears to inhibit stress, which we attribute to increased local compression of the microstructures: this reduces the amount of anodophilic creep that can occur at each microstructure, reducing the amount of stress that can be developed.

Figure 2(h) shows maximum stress and maximum contraction for microstructured PVC-DIDA gels with various mass ratios of PVC to DIDA. The highest maximum stress was achieved at one particular ratio of PVC to plasticiser, a finding that is consistent with PVC-DBA gels [20]. The electromechanical properties of microstructured PVC-DIDA gel samples were generally consistent between samples (Figure 2(i)).

In addition to functioning as single-unit soft actuators, the microstructured PVC-DIDA gels presented here can be stacked in series to form complete artificial muscles. We made a multilayer PVC-DIDA gel artificial muscle by interleaving ten microstructured PVC-DIDA gel samples with conductive fabric electrodes (Figure 3(a)). The completed muscle was highly flexible, made of soft gel and soft flexible fabric (Figure 3(b) shows the flexibility of a four-gel-layer artificial muscle). A three-gel-layer artificial muscle was capable of cyclic actuation at 0.1 Hz (Figure 3(c)), and the ten-gel-layer artificial muscle contracted by 5.5 % when 600 V was applied (Figure 3(d), Video S3). We did not explicitly investigate long-term cyclic activation with these samples, however they have delivered more than 100 cycles with no obvious degradation.

The meltability of thermoplastic electroactive gels also allows for recycling of artificial muscles. We diced a microstructured PVC-DIDA gel sample and pressed it into a new microstructured gel using a hot press (as in Figure 1(c)). The resultant recycled gel sample continued to exhibit anodophilic actuation. However, the most exciting consequence of the discovery of thermoplastic electroactive gels is the potential for fully 3D-printable electroactive gel artificial muscles.

Most low-cost 3D printers are descendants of the RepRap (replicating rapid prototyper), and fabricate parts using fused filament fabrication (FFF) [39,40], whereby material is extruded through a temperature-controlled hot-end. The most popular material, polylactic acid (PLA), has a melting temperature of 150–160 °C [40]. The melting temperature used for the electroactive gels in this article is only slightly higher (170–180 °C), suggesting their suitability for 3D printing. In addition to the gel itself, PVC gel actuators require electrodes, which could be 3D printed from a range of readily available conductive materials, such as conductive graphene PLA or elastic conductive graphene TPU (thermoplastic polyurethane).

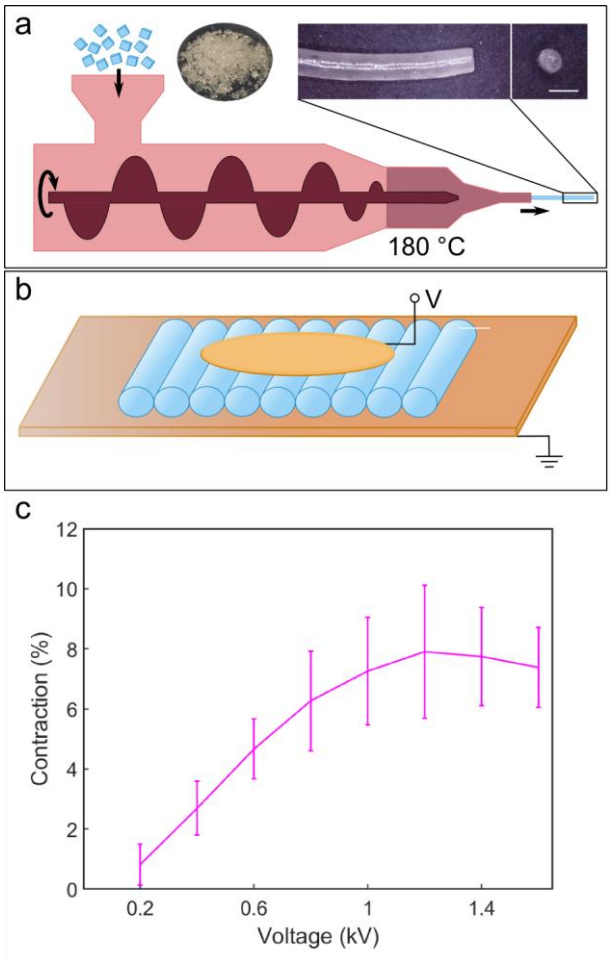

Figure 4. PVC-DIDA extruded structures. (a) Extrusion of PVC-DIDA gel. Scale bar indicates 1 mm. (b) Structure of actuator made from extruded PVC-DIDA gel. (c) Contraction as voltage is varied for an actuator made from extruded PVC-DIDA gel. Points are averages of three trials and error bars show ± one standard deviation between trials.

To assess the suitability of PVC-DIDA gel for 3D printing, we demonstrated its extrudability using a commercially available desktop extruder. We manufactured a large quantity of PVC-DIDA gel by heating precursor plastisol to around 170 °C on a hot plate. This gel was then cooled, diced into small cubes (side length roughly 10 mm) and fed into the extruder set to 180 °C, which extruded a continuous smooth PVC-DIDA gel filament (Figure 4(a)). We were able to extrude continuous PVC-DIDA gel filaments more than 3 m long using this technique, and the smallest diameter filament we extruded was 0.4 mm.

In 3D-printing applications, this extruder could be mounted on a three-axis stage to form an electroactive gel 3D printer. Alternatively, the swelled plasticised PVC gel (crumbly paste) could be used as a direct precursor, with a syringe or paste extruder propelling the precursor plastisol through a hot end.

To demonstrate how 3D-printed electroactive gel structures could be used as artificial muscles, we used the extruded PVC-DIDA gel filament to form a simple artificial muscle structure (Figure 4(b)). This extruded PVC-DIDA gel artificial muscle structure contracted by up to 11% under an applied voltage of 1.2 kV (field strength 1.04 MV m-1) (Figure 4(c)).

Conclusion

In this article, we introduced the concept of thermoplastic electroactive gels, electroactive gel materials which can be melted and reformed. We introduced an example material, PVC-DIDA gel, which can deliver actuation stress up to 2.137 kPa, contraction up to 17.75% and cyclic actuation, suitable for artificial muscle applications. Compared with other electroactive actuator technology, this contraction far exceeds the 0.1% typical for piezoelectric actuators [41] and the 10% exhibited by electrostatic actuators such as the stacked variable capacitance motor [42] and Peano-HASEL actuators [43]. It is comparable with PVC-DBA gels (14-17% contraction [7,30]) and mammalian muscle (20% contraction [41]), but falls short of the contraction of stacked dielectric elastomer actuators (46% [44]), electromagnetic linear actuators (50% [41]), and recent electro-ribbon actuators (>99% [45]). The actuation stress is of the order of the 4 kPa recovery stress reported for DBA-PVC gel [7], and requires improvement before reaching the 350 kPa reported for biological muscle [41]. Nonetheless, the current actuation stress implies PVC-DIDA gel actuators could be useful in low-force Soft Robotic applications. Examples include actuation of hair follicles or chromatophores in artificial skin, actuation of plant robots, and positioning of light components such as soft lenses.

These newly discovered thermoplastic electroactive PVC gels are excellent candidate materials for 3D printing artificial muscles. The gels described in this article exhibit impressive electroactivity, however are not yet ready for certain demanding applications, such as the replacement of failing biological muscle, which requires near-instantaneous response times and low activation voltages. However, the general concept described here can be readily applied to a wide range of gel materials, and it is expected that new artificial muscles using alternative materials could deliver improved performance characteristics. For example, hydrolysed polyacrylamide (PAAM) gels exhibit actuation under electric fields as low as 500 Vm^{-1} [47,48].

The number of plasticisers that can be used to plasticise PVC is very large, implying many new thermoplastic PVC gels warrant further study. Furthermore, other materials such as TPU gels also exhibit anodophilic behaviour [6], and therefore could be well suited as materials for 3D-printed artificial muscle structures. Some materials, such as TPU films, exhibit cathodophilic behaviour, which could also be captured for actuation [49]. Based upon the findings of this

work, a vast range of thermoplastic electroactive gels can be studied and assessed for suitability for 3D-printable artificial muscles.

Supplementary Methods

Preparation of swelled plastisol (precursor material for PVC-DIDA gels): PVC powder (346764, Sigma-Aldrich, USA) and plasticiser (diisodecyl adipate, 460214, Sigma-Aldrich, USA) were prepared in the chosen mass ratio (1:2 to 1:6) in a petri dish and mixed well by hand. This mixture was placed on a hot plate (HCB 123, Adam Equipment, UK) set to 170 °C. The mixture was stirred while heating to around 130 °C, recorded using an infrared thermometer (Fluke 62 MAX, Fluke, USA). After several minutes, the PVC particles had fully absorbed the plasticiser, and the mixture transitioned to a wet crumbly paste. This swelled plastisol was then either used immediately or stored for later use as a precursor material for PVC-DIDA gels.

Hot pressing of microstructured PVC-DIDA gels: A hot press was pre-heated to 180 °C. A negative mould (aluminium, manufactured using electrical discharge machining) was prepared by the addition of 5 g of swelled plastisol and placed inside the press. The press was then slowly closed until it reached around 1 mm from a fully closed state and held still for 10 s to pre-heat the plastisol. The press was then fully closed over the course of 20 s, after which additional pressure was rapidly applied up to 10 MPa. The press was then immediately released, and the contents (gel and mould) were rapidly cooled by placing them into a dish filled with a shallow pool of room temperature water. After cooling for 1 minute, the microstructured gel was removed from the mould using tweezers.

Adhering microstructured PVC-DIDA gels to cathodes: Cathodes, made from 2 mm thick aluminium sheet, were prepared by adding a small quantity of PVC-DIDA gel. The cathodes heated using a hot plate. Upon reaching around 170 °C, the gel began to melt and was spread evenly across the surface of the cathode. The cathode was then removed from the hot plate, and a sample of microstructured PVC-DIDA gel was placed microstructure-up on the melted gel layer on the cathode and held in place with a 300 g mass until cooled.

Isometric test setup: The isometric test setup is shown in Figure 2(a). A sample of microstructured PVC-DIDA gel was adhered to an aluminium cathode as described above and placed atop a fixed precision jack stand. Above the gel sample, an anode was attached to a fixed load cell. The degree of compression of the gel sample could be adjusted by adjusting the height of the jack stand. A laser displacement meter (LK-G402, Keyence, Japan, not shown) was used to record the height of the jack stand. To detect zero compression, the setup was initiated with a gap between the anode and the gel sample. The gel sample was slowly raised until the load cell registered a small increase in force, implying that the gel sample had contacted the load cell. The point was chosen as the zero-compression-point. To record isometric force at different levels of contraction, the jack stand was adjusted to control the compression of the gel sample. A lab computer running MATLAB was used to control a data acquisition device (NI USB-6343, National Instruments, USA), which controlled high-voltage amplifiers (5HVA24-BP1, UltraVolt, USA)

Isotonic test setup: The isotonic test setup is shown in Figure 2(b). A sample of microstructured PVC-DIDA gel was placed on a planar copper cathode. A disc-shaped, 20 mm diameter, 1 mm thick copper anode (mass 2.5 g) was placed on top of the sample. The same data acquisition device, high voltage amplifiers and laser displacement meter was used as in the isometric test setup.

Determination of initial stiffness of microstructured PVC-DIDA gels: The isometric test setup was used to determine the initial stiffness of the microstructured PVC-DIDA gels. The microstructures were each compressed by 50 μm and the force measured by the load cell compared to when the gels were uncompressed was recorded.

Fabrication of stacked PVC-DIDA gel artificial muscle: The stacked PVC-DIDA gel artificial muscle (Figure 3(a)) was made from 10 layers of microstructured PVC-DIDA gel and 11 layers of copper/tin coated conductive fabric (Zelt, Mindsets (UK) Limited, UK). The fabric was cut into 40 by 40 mm square electrodes with connector tags. Starting with an electrode, the artificial muscle was assembled by alternately placing gel samples and electrodes to form a stack.

Extrusion of PVC-DIDA gel: PVC-DIDA gel was extruded using a desktop extruder (Noztek Pro, Noztek, UK). The hot end was set to 180 °C. A sample of PVC-DIDA gel was diced into small cubes of side length roughly 10 mm and placed into the hopper. The extruder motor was then switched on, rotating the extrusion screw and propelling the diced PVC-DIDA gel down the barrel towards the hot end. A smooth continuous PVC-DIDA gel filament was extruded from the hot end.

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Data Access Statement

Data necessary to support conclusions are included in the article.

References

[1] Madden J D W, Vandesteeg N A, Anquetil P A, Madden P G A, Takshi A, Pytel R Z, Lafontaine S R, Wieringa P A and Hunter I W 2004 Artificial Muscle Technology: Physical Principles and Naval Prospects *IEEE J. Ocean. Eng.* **29** 706–28

[2] Hines L, Petersen K, Lum G Z and Sitti M 2017 Soft Actuators for Small-Scale Robotics *Adv. Mater.* **29** 1603483

[3] Pelrine R 2000 High-Speed Electrically Actuated Elastomers with Strain Greater Than 100% *Science* **287** 836–9

[4] Kim K 2002 A novel method of manufacturing three-dimensional ionic polymer–metal composites (IPMCs) biomimetic sensors, actuators and artificial muscles *Polymer (Guildf).* **43** 797–802

[5] Ionov L 2014 Hydrogel-based actuators: possibilities and limitations *Mater. Today* **17** 494–503

[6] Xia H, Hashimoto Y and Ni Q-Q 2016 Electrically Triggered Actuation of Plasticized Thermoplastic Polyurethane Gels *Macromol. Mater. Eng.* **301** 864–9

[7] Ogawa N, Hashimoto M, Takasaki M and Hirai T 2009 Characteristics evaluation of PVC gel actuators *2009 IEEE/RSJ International Conference on Intelligent Robots and Systems* (IEEE) pp 2898–903

[8] Rogovina L Z and Slonimskii G L 1974 Formation, Structure, and Properties of Polymer Gels *Russ. Chem. Rev.* **43** 503–23

[9] Ferry J D 1948 Viscoelastic properties of polymer solutions. *J. Res. Natl. Bur. Stand. (1934).* **41** 53–62

[10] Zolfagharian A, Kouzani A Z, Khoo S Y, Moghadam A A A, Gibson I and Kaynak A 2016 Evolution of 3D printed soft actuators *Sensors Actuators A Phys.* **250** 258–72

[11] Peele B N, Wallin T J, Zhao H and Shepherd R F 2015 3D printing antagonistic systems of artificial muscle using projection stereolithography *Bioinspir. Biomim.* **10** 055003

[12] Yap H K, Ng H Y and Yeow C-H 2016 High-Force Soft Printable Pneumatics for Soft Robotic Applications *Soft Robot.* **3** 144–58

[13] Wehner M, Truby R L, Fitzgerald D J, Mosadegh B, Whitesides G M, Lewis J A and Wood R J 2016 An integrated design and fabrication strategy for entirely soft, autonomous robots *Nature* **536** 451–5

[14] Rossiter J, Walters P and Stoimenov B 2009 Printing 3D dielectric elastomer actuators for soft robotics *Proc. SPIE 7287, Electroactive Polymer Actuators and Devices (EAPAD) 2009* ed Y Bar-Cohen and T Wallmersperger p 72870H

[15] Schlatter S, Rosset S and Shea H 2017 Inkjet printing of carbon black electrodes for dielectric elastomer actuators vol 1016311, ed Y Bar-Cohen p 1016311

[16] McCoul D, Rosset S, Schlatter S and Shea H 2017 Inkjet 3D printing of UV and thermal cure silicone elastomers for dielectric elastomer actuators *Smart Mater. Struct.* **26** 125022

[17] Kuhnel D T, Rossiter J M and Faul C F J 2018 3D printing with light: towards additive manufacturing of soft, electroactive structures *Electroactive Polymer Actuators and Devices (EAPAD) XX* vol 1059411, ed Y Bar-Cohen (SPIE) p 34

[18] Uddin M Z, Watanabe M, Shirai H and Hirai T 2002 Creeping and Novel Huge Bending of Plasticized PVC *J. Robot. Mechatronics* **14** 118–23

[19] Xia H, Takasaki M and Hirai T 2010 Actuation mechanism of plasticized PVC by electric field *Sensors Actuators A Phys.* **157** 307–12

[20] Ali M, Ueki T, Tsurumi D and Hirai T 2011 Influence of Plasticizer Content on the Transition of Electromechanical Behavior of PVC Gel Actuator *Langmuir* **27** 7902–8

[21] Xia H and Hirai T 2010 Electric-Field-Induced Local Layer Structure in Plasticized PVC Actuator *J. Phys. Chem. B* **114** 10756–62

[22] Yamano M, Ogawa N, Hashimoto M, Takasaki M and Hirai T 2009 A contraction type soft actuator using poly vinyl chloride gel *2008 IEEE International Conference on Robotics and Biomimetics* (IEEE) pp 745–50

[23] Park W, Bae J W, Shin E and Kim S 2016 Development of a flexible and bendable vibrotactile actuator based on wave-shaped poly(vinyl chloride)/acetyl tributyl citrate gels for wearable electronic devices *Smart Mater. Struct.* **25** 115020

[24] Furuse A and Hashimoto M 2017 Development of novel textile and yarn actuators using plasticized PVC gel *Proc. SPIE 10163, Electroactive Polymer Actuators and Devices (EAPAD) 2017* ed Y Bar-Cohen p 1016327

[25] Helps T, Taghavi M and Rossiter J 2018 Towards electroactive gel artificial muscle structures *Electroactive Polymer Actuators and Devices (EAPAD) XX* ed Y Bar-Cohen (SPIE) p 300

[26] Xia H, Ueki T and Hirai T 2009 Electrical Response and Mechanical Behavior of Plasticized PVC Actuators *Adv. Mater. Res.* **79–82** 2063–6

[27] Park W, Shin E and Kim S 2017 Enhanced Design of a Soft Thin-Film Vibrotactile Actuator Based on PVC Gel *Appl. Sci.* **7** 972

[28] Whelan T and Goff J 1990 Plasticized Polyvinyl Chloride *Injection Molding of Thermoplastic Materials - 2* (Boston, MA: Springer US) pp 126–41

[29] Harden D H and Koch R D 1967 Plastisol Coatings and Application Techniques *Orthot. Prosthetics* **21** 128–43

[30] Ali M and Hirai T 2011 Characteristics of the creep-induced bending deformation of a PVC gel actuator by an electric field *J. Mater. Sci.* **46** 7681–8

[31] Tokoro H and Hashimoto M 2014 Characteristics of a non-woven PVC gel actuator *2014 IEEE/ASME International Conference on Advanced Intelligent Mechatronics* (IEEE) pp 100–5

[32] Li Y and Hashimoto M 2015 PVC gel based artificial muscles: Characterizations and actuation modular constructions *Sensors Actuators A Phys.* **233** 246–58

[33] Kim S, Thiessen P A, Bolton E E, Chen J, Fu G, Gindulyte A, Han L, He J, He S, Shoemaker B A, Wang J, Yu B, Zhang J and Bryant S H 2016 PubChem Substance and Compound databases *Nucleic Acids Res.* **44** D1202–13

[34] Summers J W 2008 The melting temperature (or not melting) of poly(vinyl chloride) *J. Vinyl Addit. Technol.* **14** 105–9

[35] Wypych A 2013 *Plasticizers Databook* (Elsevier)

[36] Niemeyer L 1995 A generalized approach to partial discharge modeling *IEEE Trans. Dielectr. Electr. Insul.* **2** 510–28

[37] La T-G and Lau G-K 2016 Inhibiting electro-thermal breakdown of acrylic dielectric elastomer actuators by dielectric gel coating *Appl. Phys. Lett.* **108** 012903

[38] La T-G and Lau G-K 2013 Very high dielectric strength for dielectric elastomer actuators in liquid dielectric immersion *Appl. Phys. Lett.* **102** 192905

[39] Jones R, Haufe P, Sells E, Iravani P, Olliver V, Palmer C and Bowyer A 2011 RepRap – the replicating rapid prototyper *Robotica* **29** 177–91

[40] Tanikella N G, Wittbrodt B and Pearce J M 2017 Tensile strength of commercial polymer materials for fused filament fabrication 3D printing *Addit. Manuf.* **15** 40–7

[41] Hollerbach J M, Hunter I W and Ballantyne J 1991 A comparative analysis of actuator technologies for robotics *The Robotics Review* vol 2 pp 299–342

[42] Hunter I W and Lafontaine S 1992 A comparison of muscle with artificial actuators *Technical Digest IEEE Solid-State Sensor and Actuator Workshop* (IEEE) pp 178–85

[43] Kellaris N, Gopaluni Venkata V, Smith G M,

Mitchell S K and Keplinger C 2018 Peano-HASEL actuators: Muscle-mimetic, electrohydraulic transducers that linearly contract on activation *Sci. Robot.* **3** eaar3276

[44] Kovacs G, Düring L, Michel S and Terrasi G 2009 Stacked dielectric elastomer actuator for tensile force transmission *Sensors Actuators A Phys.* **155** 299–307

[45] Taghavi M, Helps T and Rossiter J 2018 Electro-ribbon actuators and electro-origami *Sci. Robot. (in Press.* **3**

[46] Rossiter J, Yap B and Conn A 2012 Biomimetic chromatophores for camouflage and soft active surfaces *Bioinspiration and Biomimetics* **7**

[47] Bassil M, El Tahchi M, Souaid E, Davenas J, Azzi G and Nabbout R 2008 Electrochemical and electromechanical properties of fully hydrolyzed polyacrylamide for applications in biomimetics *Smart Mater. Struct.* **17** 055017

[48] Bassil M, Ibrahim M and El Tahchi M 2011 Artificial muscular microfibers: hydrogel with high speed tunable electroactivity *Soft Matter* **7** 4833

[49] Watanabe M, Wakimoto N, Shirai H and Hirai T 2003 Bending electrostriction and space-charge distribution in polyurethane films *J. Appl. Phys.* **94** 2494–7